Variation of NO_2 and NO_x concentrations between and within 36 European study areas: results from the ESCAPE study

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Abbreviations:

CV: Coefficient of Variation
ESCAPE: European Study of Cohorts for Air Pollution Effects
GIS: Geographic information systems
LUR: Land Use Regression
NO_x: nitrogen oxides [µg/m³]
NO_2: nitrogen dioxide [µg/m³]
NO: nitrogen monoxide [µg/m³]
PM: particulate matter
$PM_{2.5}$: mass concentration of particles less than 2.5 µm in size
PM_{10}: mass concentration of particles less than 10 µm in size
RB: Regional Background site
SOP: Standard Operating Procedure
ST: Street site
TRAPCA: Traffic- Related Air Pollution and Childhood Asthma
UB: Urban Background site

ABSTRACT

The ESCAPE study (European Study of Cohorts for Air Pollution Effects) investigates long-term effects on human health of exposure to air pollution in Europe. Various health endpoints are analysed by using prospective cohort studies in the study areas. This paper documents the spatial variation of measured NO_2 and NO_x concentrations between and within 36 study areas across Europe.

In 36 study areas NO_2 and NO_x were measured using standardized methods between October 2008 and April 2011. In each study area 14 to 80 sites were selected, which represented a wide range of regional, urban and nearby traffic related pollution contrast. The measurements were conducted for two weeks per site in three different seasons, using Ogawa badges. Results for each site were adjusted for temporal variation using data obtained from a routine monitor background site, which operated continuously, and averaged.

Substantial spatial variability was found in NO_2 and NO_x concentrations between and within study areas. Analysis of variance showed that 40% of the overall NO_2 variance is attributable to the variability between the study areas and 60% is caused by the variability within the study areas. The corresponding values for NO_x are 30% (between the study areas) and 70% (within the study areas). The within-area spatial variability was mostly determined by the differences between traffic and urban background concentrations. The traffic/urban background concentration ratio varied between 1.09 and 3.16 across Europe. The NO_2 / NO_x ratio varied between 0.47 (Verona) and 0.72 (Heraklion) across study areas. In study areas in southern Europe the highest median concentrations were observed (Barcelona: NO_2 55 µg/m³), followed by densely populated areas in Western Europe (Ruhr area, The Netherlands). The lowest concentrations were observed in all areas in Northern Europe (e.g. Umeå: NO_2 7 µg/m³).

In conclusion, we found significant contrast in annual average NO_2 and NO_x concentration between and especially within 36 study areas across Europe. Epidemiological studies should therefore characterize intra-urban contrasts. The use of traffic indicators such as “living close to major road” as an exposure variable in epidemiological studies results in different actual NO_2 contrasts.

1. Introduction

There is now increasing evidence from epidemiological studies that exposure to ambient air pollution is associated with adverse health effects (Brunekreef and Holgate, 2002; Heinrich et al., 2004; Pope and Dockery, 2006, WHO, 2006; Rückerl et al., 2011). Adverse effects include pre-mature mortality and morbidity from cardiovascular and respiratory causes. Plausible mechanisms for these associations based upon experimental studies have been proposed, including particularly oxidative stress (Brunekreef and Holgate, 2002; Brook et al., 2010; Rückerl et al., 2011). Most studies from the USA have focussed on PM_{10} and $PM_{2.5}$ (Brook et al., 2010; Pope and Dockery, 2006). In European studies, several studies have also reported significant associations between adverse health effects and NO_2 or NO_x concentrations (Brunekreef, 2007). In these studies air pollution exposure was assessed at the residential address using dispersion models, land use regression models and traffic indicator variables. Other epidemiological studies on long-term exposure to NO_2 and other air pollutants compared the health status of populations using the contrast in city-average air pollution levels between different areas (e.g. Pope et al., 2002; Laden et al., 2006; Sunyer et al., 2006; Götschi et al., 2008). These studies generally assigned one overall average concentrations to all subjects living in each city. For NO_2 this likely results in significant misclassification as high spatial variability within urban areas has been documented previously for nitrogen dioxide (NO_2) in specifically designed studies (Lebret et al., 2000; Monn, 2001; Lewne et al., 2004). There is a substantial number of studies that have used traffic indicators as exposure variables, including distance to a major road, traffic intensity on the nearest road (HEI, 2010). A major limitation of these traffic indicators is that their value in characterizing actual air pollution exposure contrasts may differ between study areas (Jerrett et al., 2005). Some studies have made use of the spatial variation of air pollution within metropolitan areas (Gauderman et al., 2005; Gehring et al., 2006; Morgenstern et al., 2008, Jacquemin et al., 2009; Modig et al., 2009). These within-city studies often characterized air pollution with the concentration of NO_2 and NO_x obtained from either spatially dense monitoring networks, land use regression models based upon such networks or dispersion models (Jerrett et al., 2005; Hoek et al., 2008; Modig et al., 2009; Levy et al., 2010).

In 1999, the European Commission established limit values for NO_2, NO_x, PM_{10} and some other air pollutants in the Air Quality Daughter Directive 1999/30/EC (EC, 1996),

which was replaced 2008 by the new Directive 2008/50/EC on ambient air quality and cleaner air for Europe (EC, 2008). The existing air quality guidelines for NO_2 and PM_{10} are currently being exceeded at many locations throughout Europe and Germany (Giannouli et al., 2011; European Environment Agency 2006; Airbase 2007; Velders and Diederen, 2009). There is therefore substantial interest at the EU policy level in the health effects of current air pollution levels including NO_2, focussing especially on European studies.

A comparison of NO_2 concentrations measured either in study specific monitoring programme (Hazenkamp-von Arx et al., 2004) or in routine monitoring networks across Europe (e.g. Airbase data used in Beelen et al., 2009) showed significant contrast across Europe. The concentrations were generally lowest in Northern Europe and highest in the major cities and Southern Europe.

NO_2 is often used as an indicator of the complex mixture of traffic-related air pollution containing also fine and ultrafine particles. The ratio of NO_2 to other components e.g. soot in emissions of motorized road traffic has changed in the last decade (Williams and Carslaw, 2011). Specifically, the fraction of primary NO_2 emissions has increased.

In 2008 we embarked upon a European-wide study of long-term air pollution exposure health effects. The ESCAPE study - **E**uropean **S**tudy of **C**ohorts for **A**ir **P**ollution **E**ffects - assesses exposure-response relationships between long-term exposures to ambient air pollution using prospective cohort studies in 15 different European countries (http://www.escapeproject.eu).

As a key interest of epidemiological long-term studies is in within-urban variation of air pollution and the most routine monitoring networks are not sufficiently dense to characterize intra-urban concentration gradients, we decided to carry out study specific monitoring, which was independent of routine monitoring networks. In all 36 study areas, NO_2 and NO_x were measured with passive samplers. In 20 of these areas, we also measured $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10}, and PM_{coarse} (Eeftens et al., submitted). The measured average concentrations were combined with geographic predictors to develop land use regression (LUR) models (Jerrett et al., 2005; Hoek et al. 2008). In all study centres a common protocol was used to ensure high standardization of all procedures across the 36 European study areas The standardization of the measurements and the selection of the locations using a common protocol across a wide range of European

197 settings (i.e. >1000 monitoring sites across Europe) is one of the major strength of this
198 study.

199 The aim of this paper is to assess the variation of measured NO_2 and NO_x
200 concentrations between and within 36 European study areas. We further assessed the
201 variability across Europe of the increase of NO_2 and NO_x concentrations at traffic
202 stations versus urban background stations and the NO_2 / NO_x ratio. The companion
203 paper focuses on the PM measurement (Eeftens et al., submitted).

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205 **2. Methods**

206 *2.1 ESCAPE exposure assessment*

207 The objective of the ESCAPE study is to investigate relationships of long-term air
208 pollution exposure and health (for further details please refer to Eeftens et al.,
209 submitted).

210 As indicated in Figure 1 in 20 study areas both particulate matter ($PM_{2.5}$, $PM_{2.5}$
211 absorbance, PM_{10}, and PM_{coarse}) and NO_2/NO_x were measured (NO_x + PM study areas)
212 whereas in 16 remaining study areas NO_2/NO_x only was measured (NO_x only study
213 areas). In all 36 study areas, NO_2 and NO_x were measured at 40 locations spread over
214 the whole study area; whereas the PM measurements were performed at 20 sites
215 selected out of 40 sites in total (Eeftens et al., submitted).

216 Exposure assessment was highly standardized within ESCAPE. All local centres used
217 the same passive sampler, namely the Ogawa badge (htttp://www.ogawusa.com). All
218 badges were prepared and analyzed by one central laboratory at Institute for Risk
219 Assessment Sciences (IRAS), Utrecht, using the ESCAPE Standard Operating
220 Procedure (SOP) available from the ESCAPE project website
221 (http://www.escapeproject.eu/manuals/). The SOP defined the field measurement
222 procedures including Quality Assurance and Quality Control and calculations. A study
223 manual provided details on site selection. A workshop was organized for all field
224 workers to further harmonize procedures. Site selection and calculations in a centrally
225 provided Excel file were centrally checked.

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2.2 Study area and site selection

The spatial distribution of the cohort subject addresses determined the borders of the study area. Within each study area the measurement sites were selected to represent the anticipated spatial variation of air pollution at home addresses of subjects in the cohort studies. The long-term average ambient air concentration is a function of the regional background, additional pollution from all (sub)urban sources (resulting in an urban background) and pollution from local sources (e.g. traffic on nearby busy streets). In all areas street sites were overrepresented compared to the fraction of addresses on major roads, as the goal was to describe spatial variation in the area of which traffic is a main source. The requirement was to select a range of realistic traffic intensities, not only the busiest streets in the area. Other sources were also considered, e.g. specific industries, major ports. In some areas, altitude was also a factor in selecting sampling sites.

The measurement sites were classified as regional background, urban background and street sites. A street site was considered a site in a major road carrying at least 10,000 vehicles per day. Measurements were typically made at the façade of the homes, as we were interested in characterizing residential exposures. We hence did not measure at the kerbside. An urban background site was defined as a site with fewer than 3,000 vehicles per day passing within a 50m radius. The distinction between regional and urban background was not strictly defined in the study manual, but typically involved measurements in the smaller towns of the cohort.

Based on the ESCAPE guidelines each local research center made a site selection proposal with a detailed characterization of the sites including Google maps of the study area and sites. The proposal was discussed by the ESCAPE exposure working group to harmonize the site selection across the centers. Each selected site was repeatedly geocoded using a GPS (e.g. at each start of a measurement). Because accuracy is very important – spatial variability of air pollution concentrations occurs within tens of meters from major roads – the measured geocodes were plotted and checked on GIS maps with high resolution (e.g. road network, building ground map) and if necessary the geocodes were corrected to the original spot of measurement.

An overview of the characteristics of study areas is presented in Table 1 and in more detail in Online Supplement A.

2.3 Sampling and analysis

The standard operating procedure SOP is available from the ESCAPE project website (http://www.escapeproject.eu/manuals/). NO_2 and NO_x were measured using Ogawa diffusion badges. A detailed description of sampling and analysis has been published previously (van Roosbroeck et al., 2006). The sampler contains two collection filters that are coated with a reactive chemical, one for sampling NO_2 and one for sampling NO_x (NO_2 plus NO). NO is calculated by subtraction. Ogawa badges were transported from the central laboratory in individual plastic bags and cooled during transport and storage. The analysis is spectrophotometrically based upon the Saltzman method (van Roosbroeck et al., 2006). The preparation and chemical analysis of all Ogawa badges was performed centrally in one laboratory (IRAS, Utrecht). From each batch of 40 filters obtained from the manufacturer, four filters were kept at IRAS laboratory as lab blanks. These four lab blanks were analyzed on the same day as the exposed filters and their results were subtracted from results of filters in the same batch.

To establish the agreement with the chemiluminescence method being the European reference method, we compared the Ogawa diffusion badges during every 2-week sampling period with a chemiluminescence monitor in each study area. The Ogawa samplers were located in direct vicinity to the chemiluminescence monitor. The results will be published soon in a separate publication.

2.4 Sampling campaign

Because of limited equipment availability, especially for PM measurements, the study areas were divided into a first and second year group (Table 1). The sampling period for the first year group was between October 2008 and February 2010, and for the second year group was between November 2009 and April 2011. At each monitoring site three measurement periods of two weeks were conducted; in the cold, warm and one intermediate temperature season. The two week sampling periods were performed in weeks with no unusual events such as bonfires and major holidays (i.e. school holidays of a week or longer).

In the NO_x only study areas the two week measurements were done almost simultaneously at all sites – start day of all measurements was within a time period of maximum three days. In contrast, due to equipment limitation for PM sampling, the measurements in the NO_x + PM study areas were performed simultaneously only at 5

sites and one continuous reference site. Thus, for completion of one measurement period four rounds of two week measurements were necessary. Each group of 5 sites included different site types, e.g. regional, urban background and traffic. NO_2 and NO_x in the NO_x + PM study areas was measured exactly at the same time as the PM measurements were performed.

The reference site was used for sampling during all sampling periods, covering a full year. This reference site was located at an urban or rural background location and measured over the whole year. The site was used to adjust for temporal variation. If a PM and/or NO_x measurement failed, the measurement was repeated, preferably in the same season.

2.5 Adjustment for temporal variation

Air pollutants have a substantial temporal variation. Therefore adjustment for temporal variability is essential if annual means of the measured NO_2 and NO_x concentrations are calculated, particularly for the NO_x + PM areas with non-simultaneous measurements. For details we refer to the PM companion paper (Eeftens et al., submitted). Briefly, the difference between the concentration for a specific two week sampling period and the annual average at the reference site was subtracted from each measurement. This procedure is based upon procedures developed previously within the TRAPCA (Traffic-Related Air Pollution and Childhood Asthma) study (Hoek et al., 2002). The annual average of NO_2 and NO_x is calculated from the ESCAPE reference site for the NO_x + PM areas. For the NO_x only sites, an urban or rural background station routine monitoring data was used. We did not require a year-long reference campaign, because measurements were already made simultaneous and only occasional missing data could lead to bias in the spatial comparison of calculated averages. In Umeå where no appropriate routine monitoring site was available, adjustment was made using the average of the three sampling campaigns of all sites with complete data. This approach only takes care of bias due to missing data and does not scale the data to an annual average.

In the following this paper focuses on adjusted annual average NO_2 and NO_x concentration. We report concentrations in $\mu g/m^3$. This has implications e.g. for reported NO_2 / NO_x ratio. In case of equal NO_2 and NO concentrations in ppb, the NO_2 / NO_x ratio is 0.50 expressed in ppb and 0.60 in $\mu g/m^3$. If the NO_2 is twice the NO concentration in ppb, the NO_2 / NO_x ratio is 0.67 expressed in ppb and 0.75 in $\mu g/m^3$.

2.6 Quality control

Four field blanks and four duplicates were collected in each of the three measurement periods for the NO_x only study areas, to achieve a total of 12. For the NO_x + PM areas in each sampling period one field blank and duplicate were taken, which resulted in at least 12 field blanks and duplicates. Typically more blanks and field duplicates were taken as these samples were taken in each sampling period of the year-round reference site. The plastic bags of the field blanks were opened at the measurement spot for a short time. Duplicates were installed at the same location. The limit of detection was calculated as three times the standard deviation of the blanks. Precision was calculated from field duplicates according to Eeftens et al. (submitted).

2.7 Data analysis

Locally calculated adjusted annual averages were gathered centrally and their range and distribution were calculated, and stratified by site type. To quantify the amount of spatial variation relative to the background level, the interquartile range and total range (maximum-minimum) were calculated as a percentage of the mean. For each study area, we used ANOVA (SAS 9.2, PROC GLM) to test for significant differences between urban background and street sites and (where applicable) between urban and regional background sites. We also tested if urban background levels differed significantly between study areas. The same analyses were performed for street sites and regional background sites (where applicable), again using SAS 9.2, PROC GLM. Correlation R^2-values between components were determined using SAS 9.2, PROC REG. Percentages of between and within area variance were determined using analysis of variance with PROC VARCOMP, METHOD=reml.

3. Results

ESCAPE included study areas across Europe that were of substantially different size (Figure 1, Table 1 and Online Supplement A). Please note that the study areas in Table 1 are sorted from the North to the South. We kept this order in all following tables and figures. The size of the areas is given by the distribution of the included cohorts. Some areas were restricted to a single city (e.g. Rome, Grenoble, Erfurt) or a metropolitan area including more rural / suburban areas (Stockholm country, Athens). The study also included large study areas such as the Ruhr area, Catalunya and the Netherlands /

Belgium. The included main cities were also of very different size, ranging from Umeå (~100,000 inhabitants) to the largest European metropolitan areas such as Paris and London with several million of inhabitants. Due to the differences between the study areas the number of regional, urban background and street sites in each study area is also different.

3.1 Quality control

Detection limits were low for NO_2 for all study areas and very few samples were below the limit of detection (Table 2). Detection limits were slightly higher for NO_x but in nearly all study areas very few samples were below the limit of detection. Only in Turin, Catalunya and Albacete a sizable fraction was below the detection limit. This was partly due to unexplained outliers which were all included in the detection limit calculations presented in Table 2. Samples below the detection limit were retained at their original value.

Precision expressed as the Coefficient of Variation (CV) calculated from field duplicates (Eeftens et al., submitted) was lower than 10% in 31 areas for NO_2 and in 34 areas for NO_x (out of 36). The average CV was 9.2% for NO_2 and 6.1 for NO_x, respectively. The areas with the largest CV (especially for NO_2) were Grenoble, Oslo, Granada and Huelva. In Granada only three and in Huelva two duplicates were obtained. Precision was in 24 study areas poorer for the NO_2 measurements compared to NO_x, but the difference was mostly only minor.

3.2 Spatial contrast of average NO_2 and NO_x concentrations

Unadjusted average concentrations and average concentrations adjusted for temporal variation were very highly correlated (Online Supplement B, Table B1). Squared correlations (R^2) were generally above 0.95, indicating that temporal adjustment resulted only in small changes to the calculated average concentrations. For the NO_x only areas, this reflects that sampling was performed simultaneously and that there were few measurements that failed due to stolen badges or sampling and analysis errors. For sites with three valid observations, the adjustment involves the same scaling to an annual average. For the sites with less than three a different scaling may occur, e.g. if the winter measurement has failed, the adjustment to the average is typically larger than for the sites with valid measurements in the three seasons. For the NO_x + PM study areas, where measurements were not conducted simultaneously, measurements of two-

week samples in three seasons were apparently sufficient to obtain a fairly stable estimate of spatial contrasts, in agreement with findings of the TRAPCA study using the same approach (Lewne, 2004).

The distribution of adjusted annual average concentrations for NO_2 and NO_x is shown in Figures 2a and 2b as well as in Online Supplement B, Tables B2 and B3.

3.3 Spatial variability within study areas

In many study areas the contrast of individual averages within the study area was as large as the contrast in median concentration across the study areas (Figures 2a and 2b). Substantial spatial variability was found for NO_2 and NO_x concentrations within study areas (refer also to Online Supplement B, Tables B2 and B3). The average range for NO_2 (difference between the highest and the lowest annual average in one specific study area) was 54 $\mu g/m^3$. The lowest range was observed in Kaunas, Gyor and Bradford NO_x (28 $\mu g/m^3$) as well as in Heraklion (29 $\mu g/m^3$). The largest spatial variability for NO_2 was found in Catalunya (97 $\mu g/m^3$) followed by London and Barcelona (95 $\mu g/m^3$). The average range for NO_x was 136 $\mu g/m^3$. The lowest NO_x range was measured again in Heraklion (44 $\mu g/m^3$) followed by Huelva (58 $\mu g/m^3$) and Munich (72 $\mu g/m^3$). The largest spatial variability for NO_x was observed in Marseille (254 $\mu g/m^3$) followed by Verona (251 $\mu g/m^3$), London (239 $\mu g/m^3$) and Paris (236 $\mu g/m^3$).

The results of a restricted maximum-likelihood variance component analysis (SAS PROC VARCOMP, METHOD=reml) show that the within study areas variance of NO_2 (60.1% of the total variance) is considerable larger than the variance attributable to the differences between the study areas (39.9% of the total variance). The difference for NO_x is even more pronounced; variance component for within study area variability is 1005.5 (70% of the total variance), and for between study variability 430.2 (30% of the total variance).

The type of the measurement sites (regional background, urban background and street sites) plays also an important role in the overall variability of NO_2 and NO_x. After including the variable “type of the measurement site” to the variance analysis the results show that 30.4% of the overall NO_2 variance is attributable to the variability between the study areas, 37.4% of the variability is caused by different site type and 32.3% could be traced to the variability within the site types. The corresponding values for NO_x are

23.4% (variability between the study areas), 36.5% (variability because of different site type) and 40.1% (variability within the site types).

The distribution of annual averages of NO_2 and NO_x concentrations by site type, for each study area is shown in Figures 3 and 4, respectively. The NO_2/NO_x concentrations at urban background sites are in general lower than at street locations. The range of NO_2/NO_x concentrations is lowest for regional background sites, followed by urban background sites, whereas the largest variability across the sites was observed for street sites.

Table 3 presents the ratio of concentrations measured at regional versus urban background and the ratio of traffic to urban background sites for NO_2 and NO_x, respectively. In almost all study areas the differences between the site types were statistically significant ($p<0.05$ level).

In all study areas, the concentrations at traffic stations were higher than at urban background stations; the street site vs. urban background site ratio was 1.63 for NO_2 and 1.93 for NO_x on average for all sites over all study areas. The ST / UB ratio ranged substantially from 1.09 to 3.16 for NO_2. High ratios were found especially in Umeå, Oslo, Mideast Spain and Granada. In all areas the NO_x ST / UB ratio was larger than for NO_2. The NO_x ST / UB ratio ranges from 1.14 in Lugano to 4.24 in Umeå and 2.77 in Paris.

NO_2 concentrations were lower at regional background sites compared to urban background concentrations; the mean ratio for all sites over all study areas was 0.63 for NO_2 and 0.60 for NO_x, respectively. However, the RB / UB concentration ratio varied substantially across study areas, from 0.24 in London to 0.91 in Gyor (Table 3). Low ratios were found in London, Umeå and Mid-East Spain. The highest ratios were found in the Ruhr area and Gyor. The pattern for NO_x was similar.

3.4 Spatial variability across Europe

Because the overall mean concentration may be affected by differences in the fraction of different site types, the direct comparison of the results between the study areas (across Europe) is based mainly on urban background locations.

Substantial differences in annual average NO_2 and NO_x concentration across Europe were found (Online Supplement B, Tables B2 and B3). The lowest annual average NO_2 concentration was found in Umeå, a medium sized city in Northern Sweden.

Concentrations in the large North-European cities (Helsinki, Stockholm, Oslo, and Copenhagen) were relatively low, but similar to the smaller cities in southern (Heraklion, Crete) and central Europe (Erfurt, Gyor, Kaunas). The highest concentrations were measured in the Mediterranean area, especially Barcelona and Turin. The pattern for annual NO_2 levels measured on street locations follows more or less the pattern observed for the urban background locations, except for Paris, Lyon and Granada. The largest differences between the NO_2 concentrations measured on street sites and urban background sites were observed for Paris (31 $\mu g/m^3$) and Mediterranean areas such as Granada, Turin, Lyon, Barcelona and Catalunya (>31 $\mu g/m^3$), whereas the lowest difference between the two site types were detected for Lugano, Kaunas and Bradford.

The pattern for NO_x was similar to that of NO_2.

3.5 $NO_2 - NO_x$ relationship

In all study areas a high correlation between NO_2 and NO_x was found (Table 4). Table 4 presents also the average NO_2 to NO_x ratios measured in rural, urban background and street sites in each study area. The average NO_2 to NO_x ratio ranged from 0.51 to 0.72 for urban background sites in the different study areas. The lowest ratios were found in Verona, Geneva and Basque Country. The highest ratio was found in Copenhagen. At traffic stations the NO_2 to NO_x ratio ranged from 0.42 to 0.73, with the highest value found in Heraklion. Overall there was no clear North-South gradient in the ratio. Mean ratios at urban background sites were 0.62, 0.63 and 0.61 for the Northern-European, central-western and southern European areas, respectively.

In most areas, the NO_2 to NO_x ratio was smaller at street sites than at urban background sites. On average, the ratio was 0.69, 0.62 and 0.54 for regional background, urban background and street sites respectively. Without Mid-East Spain the ratio was 0.65 for regional background. The difference in ratio varied significantly across Europe, e.g. in Copenhagen, Paris and the Netherlands the ratio was much lower at street sites than at urban background sites, whereas in e.g. Basel, Geneva there was little difference, possibly reflecting differences in primary NO_2 emissions across Europe.

4. Discussion

Overall we found significant contrast in annual average NO_2 and NO_x concentration between and especially within 36 study areas across Europe. NO_2 concentrations at street locations were on average between 1.22 and 3.6 times higher than at urban background stations. The NO_2 / NO_x ratio varied between 0.47 and 0.72 across study areas. Concentrations were generally lower in Northern than in Southern Europe.

4.1 ST / UB contrast

The ST / UB contrast observed in our study for NO_2 and NO_x concentrations was higher and more consistent than for the particle metrics across all study areas (Eeftens et al., submitted). For $PM_{2.5}$, $PM_{2.5}$ absorbance (soot), PM_{10} and PM_{coarse} we found ratios of 1.14, 1.38, 1.23 and 1.42 respectively. This was not due to differences in the included study areas, as the NO_2 and NO_x ST/UB ratio in the areas where PM was measured were 1.54 and 1.80 respectively. Such pronounced contrast between traffic and background environments was observed also for other traffic related pollutants, including ultrafine particles (Cyrys et al., 2003a; Tuch et al., 2006; Puustinen et al., 2007; Cyrys et al., 2008; Hoek et al., 2011). Furthermore, the spatial variation of NO_2 is more comparable to the spatial variation of other traffic related primary pollutants (soot) and less to those of partly secondary pollutants such as $PM_{2.5}$ or PM_{10}. As shown in the companion paper for $PM_{2.5}$, PM_{10} and $PM_{2.5}$ absorbance (Eeftens et al., submitted) the R^2 between NO_2 and $PM_{2.5}$ absorbance (as indicator for traffic related soot) is generally high: 0.80 (range 0.55-0.91), while the correlation between NO_2 and $PM_{2.5}$ is much lower: R^2= 0.50 (range 0.02-0.90). The contrast between regional and urban background sites was more consistent and higher than observed in the companion paper for $PM_{2.5}$, PM_{10} and absorbance in almost all areas (Eeftens et al., submitted). Also the temporal variation of NO and NO_2 concentrations is often strongly correlated with those of other traffic related air pollutants, such as CO, Black Carbon and ultrafine particles (Cyrys et al., 2003b; Hagler et al., 2009; Sabaliauskas et al., 2012). Moreover, nitrogen oxides have been found to be the most important predictor variables for ultrafine particles in the urban air (Paatero et al., 2005). It reveals the role of nitrogen oxides as marker for traffic related air pollutants. Due to the close temporal correlation with other combustion related pollutants NO_2 has been often used in epidemiological studies as a marker for traffic exhaust. Its concentration is measured easily and relatively cheaply,

but one should keep in mind that it serves only as a surrogate for a set of sources and resulting mixture of air pollutants.

4.2 Variability within study areas

The observation of high within study area variability of NO_2 concentrations compared to between study area variability agrees with previous observations in four urban areas cities in western and central Europe (Lebret et al., 2000), 16 cities of the European Community Respiratory Health Survey (ECRHS) (Götschi et al., 2008) and 8 areas of the longitudinal Swiss cohort study on air pollution and lung and heart disease in adults in Switzerland (SAPALDIA) (Liu et al., 2012).

It raises one of the central questions of air pollution epidemiology. In epidemiological studies evaluating the health effects of long-term exposure to air pollutants, the place of residence explains much of the exposure contrasts between persons. Vast majority of long-term epidemiological studies compare the health status of populations living in different cities (Pope and Dockery, 2006; Götschi et al., 2008). These studies generally assigned one overall average concentrations to all subjects living in each city and use the contrast in city-average air pollution levels between different areas. This approach assumes that a limited number of monitors per area (or even only one monitor per city) could provide an unbiased estimate of the average community exposure to background pollution. The observation of large within study area contrasts which exceed between study areas contrasts raises considerable doubts about this type of analysis. For air pollutants showing pronounced spatial variability and affected by local sources (such as traffic) assessing exposure at the community level could lead to substantial misclassification and together with other factors may introduce considerable random error in the estimation of the true individual exposure or even contribute to the observed null-findings (Götschi et al., 2008). The large spatial variation of the concentration levels of traffic related air pollutants across the cities suggests that it is virtually impossible to characterize the city-average concentration with one monitoring site. Because of this, modelling of concentrations for traffic related air pollutants could be a reasonable option for exposure assessment. For the ESCAPE study we will develop study-area specific LUR models.

The observed pronounced within-area spatial contrast for NO_2 and NO_x is in a large part attributable to the differences between street and urban background sites. The ratio of the ST / UB concentrations varied widely across the study areas. The further implication

549 of this variability for epidemiological studies is that the use of traffic indicator variables
550 such as living close to a major road presents a different contrast in actual air pollution
551 exposure in different cities. This likely contributes to the observed heterogeneity of
552 estimated health effects in studies using traffic indicators (Vardoulakis et al., 2003;
553 Heinrich et al., 2004).

554 The relatively high ST / UB ratios in Northern European cities, particularly in Umeå,
555 are likely due to the low urban background concentrations in these countries. The
556 variability in ratios is probably explained by a combination of the following factors:
557 traffic intensity, fraction of heavy duty vehicles, emission factor of the car fleet related
558 to e.g. age and fuel type of cars and possibly street configuration. In interpreting the
559 (often modestly increased) magnitude of the ratio of concentrations measured at street
560 sites and urban background sites, it has to be taken into account that measurements were
561 not made at kerbsides, as is often the case in routine monitoring network street sites.
562 The average ratio further reflects an average street, not the busiest street in the area.
563 Figures 3 and 4 illustrate that there is significant variability across street sites.

564 The RB / UB ratios for NO_x were more consistent and higher than observed in the
565 companion paper for $PM_{2.5}$, PM_{10} and absorbance in almost all areas (Eeftens et al.,
566 submitted). PM_{10} and $PM_{2.5}$ have a high regional background concentration and local
567 sources increase concentrations only modestly, however absorbance as marker for soot
568 is strongly affect by local sources including traffic. The higher contrast for NO_2
569 compared to soot could be related to the differences of atmospheric lifetime for NO_x and
570 soot. Soot is enriched in submicrometer particle fraction having a longer atmospheric
571 lifetime and could be accumulated there. Higher emissions of NO_x from road traffic or
572 other combustion sources compared to the background could also play a role. The
573 slightly higher ratio of traffic/urban background for NO_2 compared to soot argues in
574 favour of this explanation, though this pattern is less clear than the urban-rural contrast.
575 The use of after treatment technology that traps soot from diesel vehicles but increases
576 NO_2 because of catalytic oxidation of collected soot may have contributed to this (Grice,
577 2009; Williams and Carslaw, 2011).

578 At 323 sites (out of 1485) in 31 study areas (out of 36), the annual average NO_2
579 concentration measured at a site exceeded 40 $\mu g/m^3$, the EU air quality guideline for the
580 annual average (in Catalunya at 50 sites, Barcelona at 33 sites, and Turin at 31 sites).
581 The comparison with the annual average guideline is limited because we sampled only

582 during three 2-week periods. However, a reference site was used to adjust all measured
583 concentrations to an annual average concentration.

584 *4.3 Contrasts across Europe*

585 The general pattern across Europe agrees with previous studies based upon study
586 specific monitoring programme (Hazenkamp-von Arx et al., 2004) or upon routine
587 monitoring stations (Beelen et al. 2009). The high NO_2 concentrations in Southern
588 European countries could be due to high traffic intensity, a large fraction of diesel-
589 powered vehicles and a higher conversion of NO to NO_2 because of relatively high
590 temperatures and ozone concentrations. Alternatively, the more densely built
591 Mediterrean cities could lead to higher concentrations. High air pollution
592 concentrations in Turin and the Po-valley in general have been reported before
593 (Hazenkamp-von Arx et al., 2004). Turin is located mainly on the left bank of the Po
594 River and surrounded by the Alpine arch. It favors thermal inversions, characterized by
595 low surface wind speeds (stagnation of the air) and trapping air pollutants in the lower
596 layers of the atmosphere. In general these unfavorable meteorological conditions are
597 characteristic for the whole Po Valley al all. The combination of the stagnant air
598 conditions with high emissions due to heavy traffic and high population density is the
599 cause of a large number of days exceeding air quality standards and some very strong
600 pollution episodes in this area (Minguzzi et al., 2005; de Meij et al., 2009; Lonati et al.,
601 2010; Fattore et al., 2011). Some of the variability across the study areas is probably
602 also due to weather variability between the two years.

603 Similar North-South gradient across Europe was also observed for $PM_{2.5}$ and PM_{10}
604 (Hazenkamp-von Arx et al., 2004; Eeftens et al., submitted). However in the NO_2
605 pattern a city size is also clearly visible. NO_2 concentrations in the moderately sized
606 cities of Heraklion, Crete, Gyor and Kaunas were similar to those in the Northern large
607 cities of Helsinki, Stockholm and Oslo, whereas the PM_{10} and $PM_{2.5}$ concentrations
608 were substantially lower in the Northern cities. This reflects the larger impact of long
609 range transported secondary aerosol on the $PM_{2.5}/PM_{10}$ levels and, in contrast, the
610 predominant role of local source emissions on the NO_x concentrations. Another example
611 is that NO_2 concentrations in London exceeded concentrations in the Netherlands and
612 the Ruhr area whereas the $PM_{2.5}$ and PM_{10} concentrations showed the opposite pattern.

613 By the comparison of the NO_2 and NO_x concentrations across the study areas one
614 should keep in mind that the study areas are of substantially different size and type

615 (which is given by the different distribution of the existing study cohorts). Some study
616 areas consist of one single city, some include more rural and/or suburban locations and
617 some others cover whole regions (Catalunya) or even countries (the Netherlands /
618 Belgium). Also the included cities differ regarding their size from small cities like
619 Umeå to the largest European metropolitan areas such as Paris or London. Because of
620 those differences also the number of regional, urban background and street sites in each
621 study area is different.

622 *4.4 NO_2 to NO_x ratio*

623 The major sources of NO_2 and NO_x are motorized road traffic, industry, shipping and
624 heating. Nitrogen oxides are emitted as NO and NO_2. In the atmosphere NO reacts with
625 ozone to form secondary NO_2. The observed NO_2 to NO_x ratio varied between study
626 areas and site types within study areas. The typically higher ratios observed at urban
627 background sites reflect enhanced transformation of NO to NO_2 through equilibrium
628 reactions with ozone at those sites. The relatively modest difference between traffic and
629 urban background sites may reflect the increase in primary NO_2 emissions close to street
630 sites. Recent studies reported evidence of increasing NO_2 to NO_x ratio from road traffic
631 emissions due to an increase in primary NO_2 emissions observed in several urban areas
632 in Europe. Primary NO_2 emissions have therefore gained importance compared to the
633 ozone / NO_x equilibrium (Keuken et al., 2009; Mavroidis and Chaloulakou, 2011). This
634 increase have been attributed to the more common use of diesel-fuelled vehicles, since
635 they emit a higher fraction of NO_2 compared to gasoline-fuelled vehicles (Grice et al.,
636 2009; Anttila et al., 2011; Carlsaw et al., 2011). In addition, the after-treatment devices
637 (such as oxidation catalyst) implemented for reducing particulate matter emissions by
638 diesel vehicles contribute to increasing fraction of primary NO_2 in NO_x (Williams and
639 Carslaw, 2011; Mavroidis and Chaloulakou, 2011). For diesel-fuelled vehicles equipped
640 with catalytic diesel particulate filters, primary NO_2 fractions of around 40 - 50% are
641 reported (Carlsaw et al., 2007).

642 Significant variability of the fraction of primary NO_2 in traffic emissions across Europe
643 was reported in the studies, but there is less information on NO_2 / NO_x concentration
644 ratios.

645 Due to higher ozone levels and a higher percentage of diesel vehicles in the car fleet, we
646 expected higher NO_2 to NO_x ratio in southern Europe. We did not find this, possibly
647 because of more street canyons within Southern European cities, in which due to poor

dispersion ozone may become a limiting factor (Vardoulakis et al. 2003). For Athens, it was suggested that the fraction of primary NO_2 is not increasing (as is the case for other urban areas in Europe) as diesel passenger cars are not allowed there and particle after-treatment technologies are not applied in Greece (Mavroidis and Chaloulakou, 2011).

5. Conclusion

We found significant contrast in annual average NO_2 and NO_x concentration between and especially within 36 study areas across Europe. NO_2 concentrations at traffic stations were on average between 1.22 and 3.6 times higher than at urban background stations. The NO_2 / NO_x ratio varied between 0.47 and 0.72 across study areas. Concentrations were generally lower in Northern than in Southern Europe, but a clear impact of city size was also found.

Epidemiological studies should therefore characterize intra-urban contrasts. The use of traffic indicators such as “living close to major road” as an exposure variable in epidemiological studies results in different actual NO_2 contrasts.

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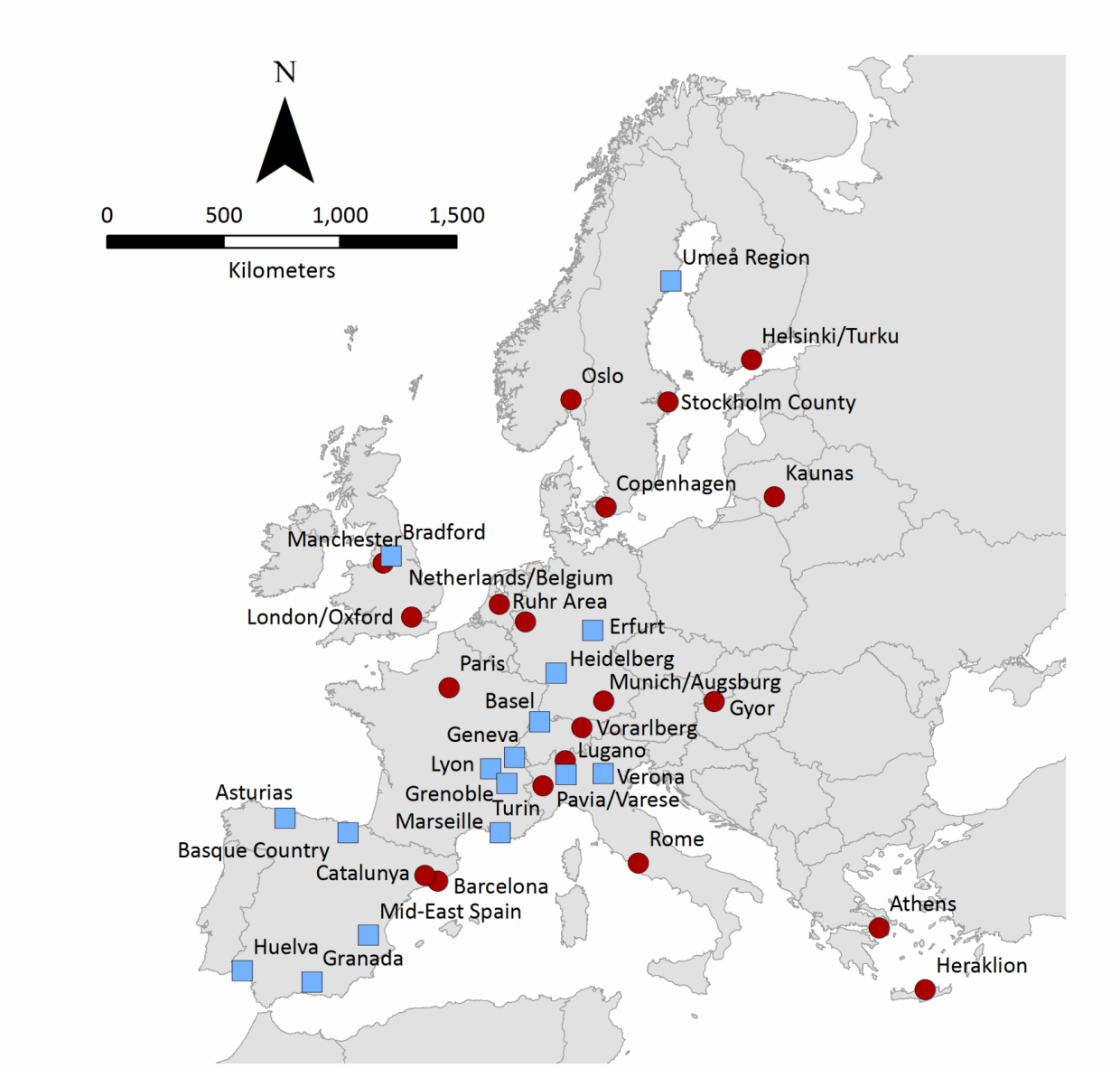


871
872
873 Figure 1: ESCAPE study areas; dark circles mark the study areas where both PM and
874 NO_x were measured. Blue squares indicate the study areas where only NO_x was
875 measured

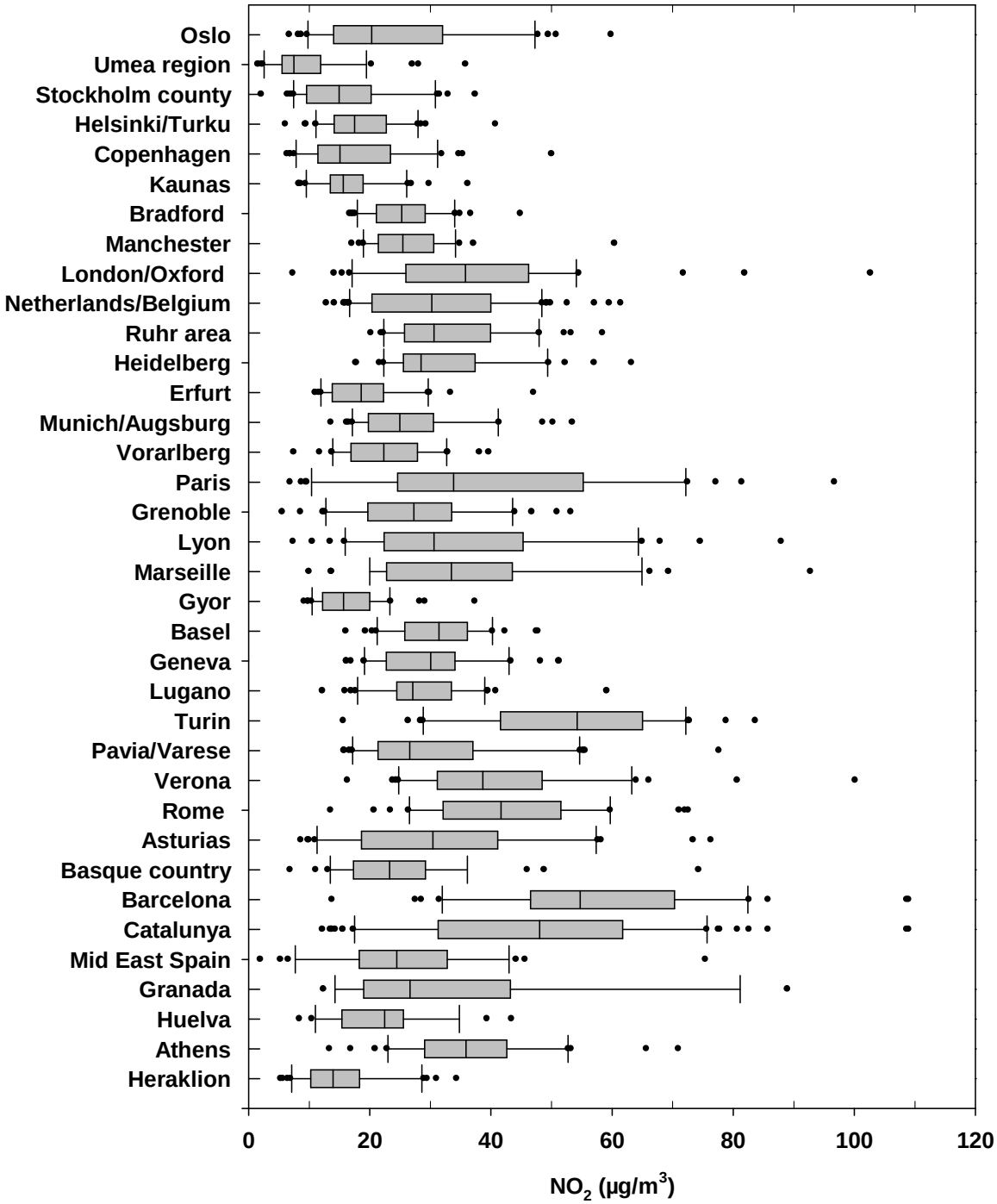


Figure 2a: Distribution of annual average concentration of NO_2 for each study area separately. Median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points.

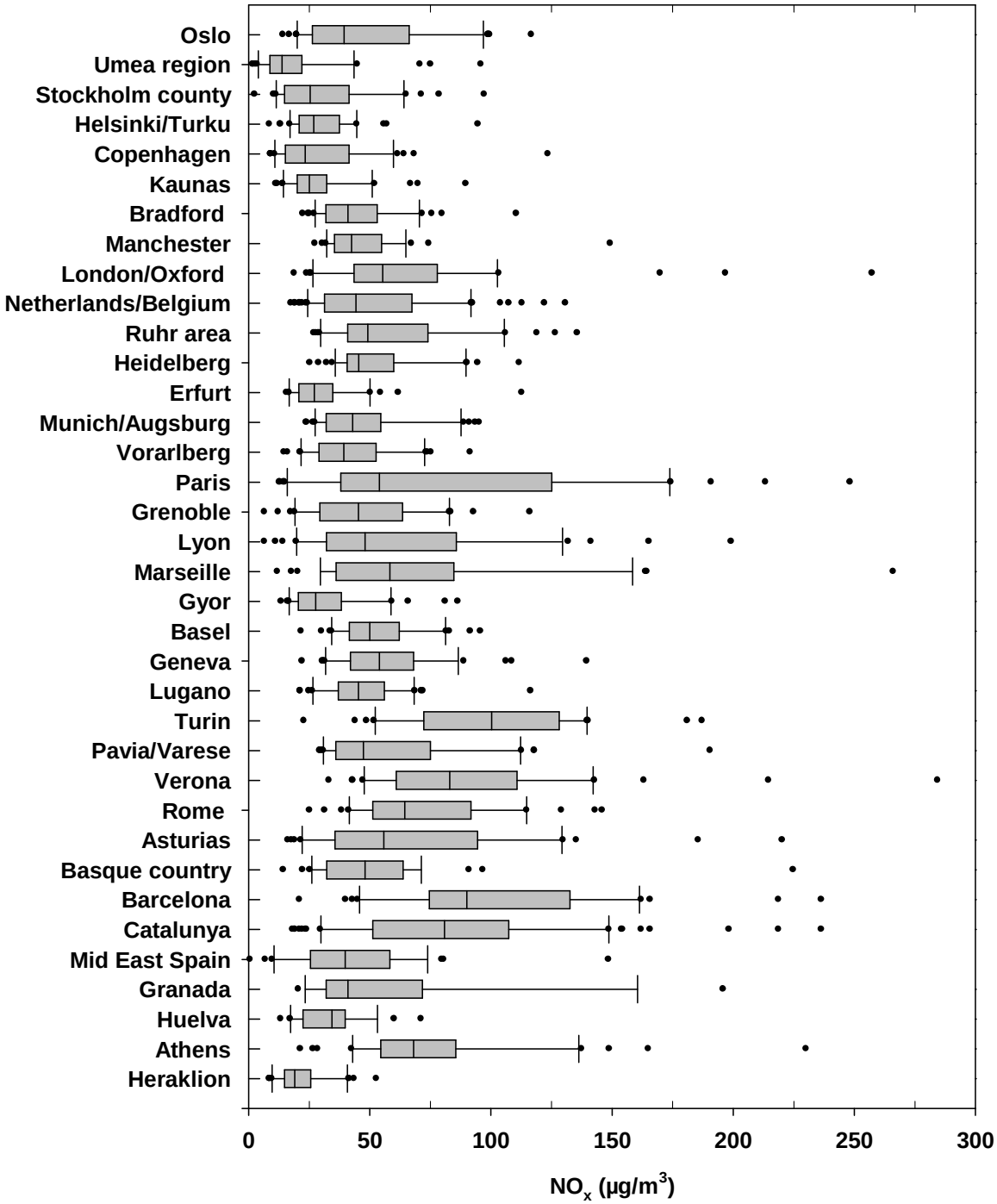
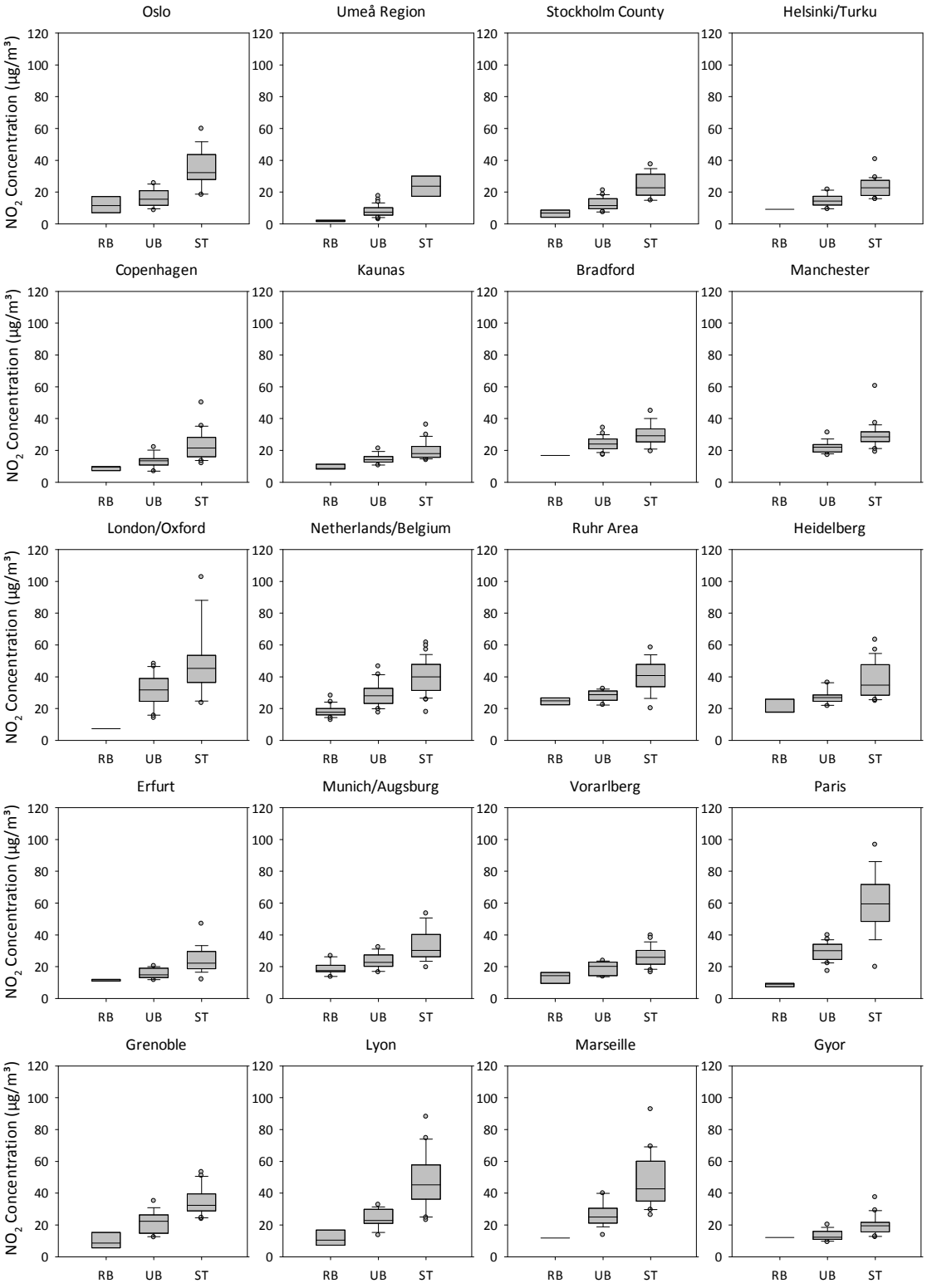


Figure 2b: Distribution of annual average concentration of NO_x for each study area separately. Median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points.


Oslo
Umeå Region
Stockholm County
Helsinki/Turku
Copenhagen
Kaunas
Bradford
Manchester
London/Oxford
Netherlands/Belgium
Ruhr Area
Heidelberg
Erfurt
Munich/Augsburg
Vorarlberg
Paris
Grenoble
Lyon
Marseille
Gyor
NO2 Concentration (µg/m³)
0
20
40
60
80
100
120
RB
UB
ST

887

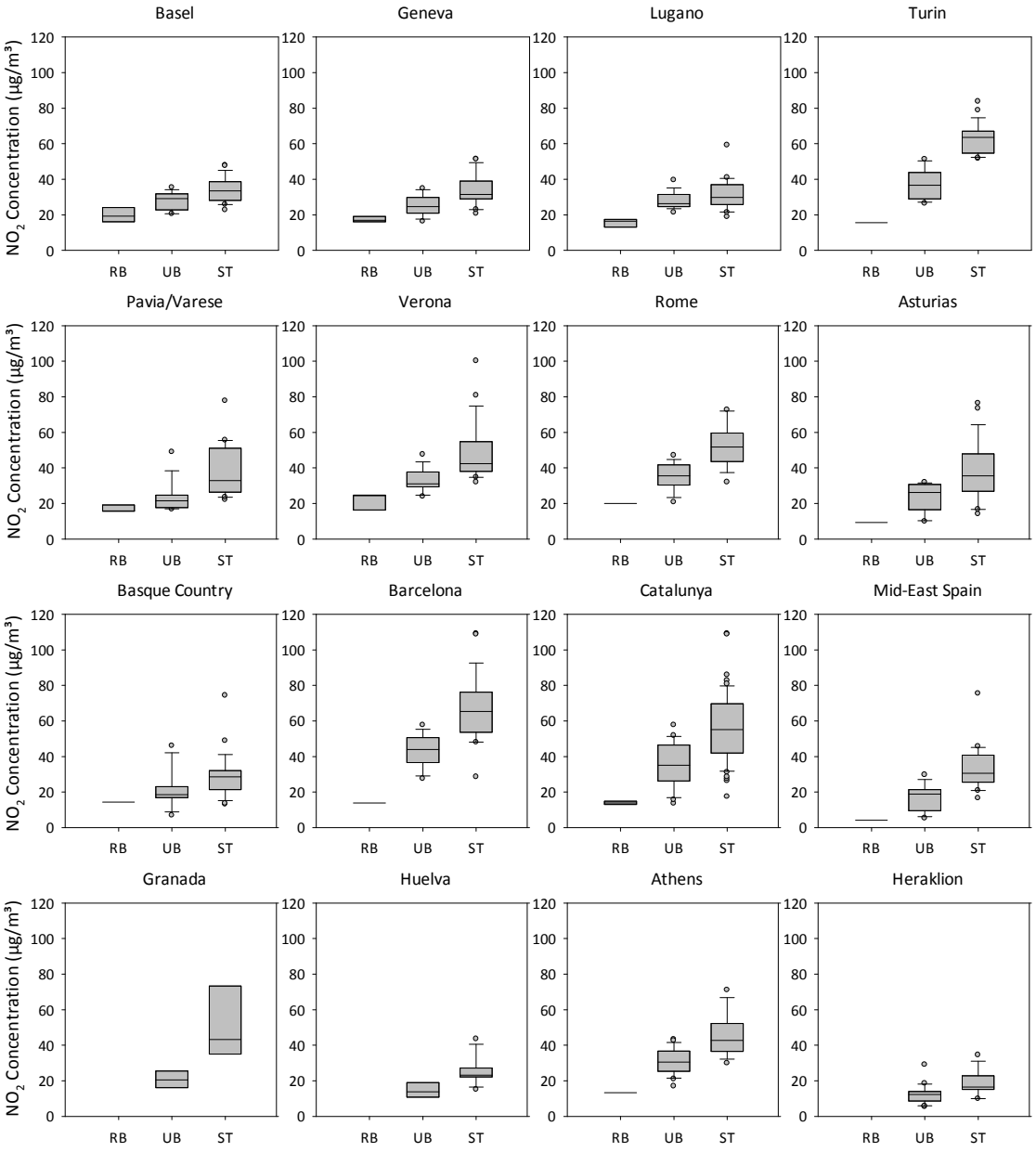
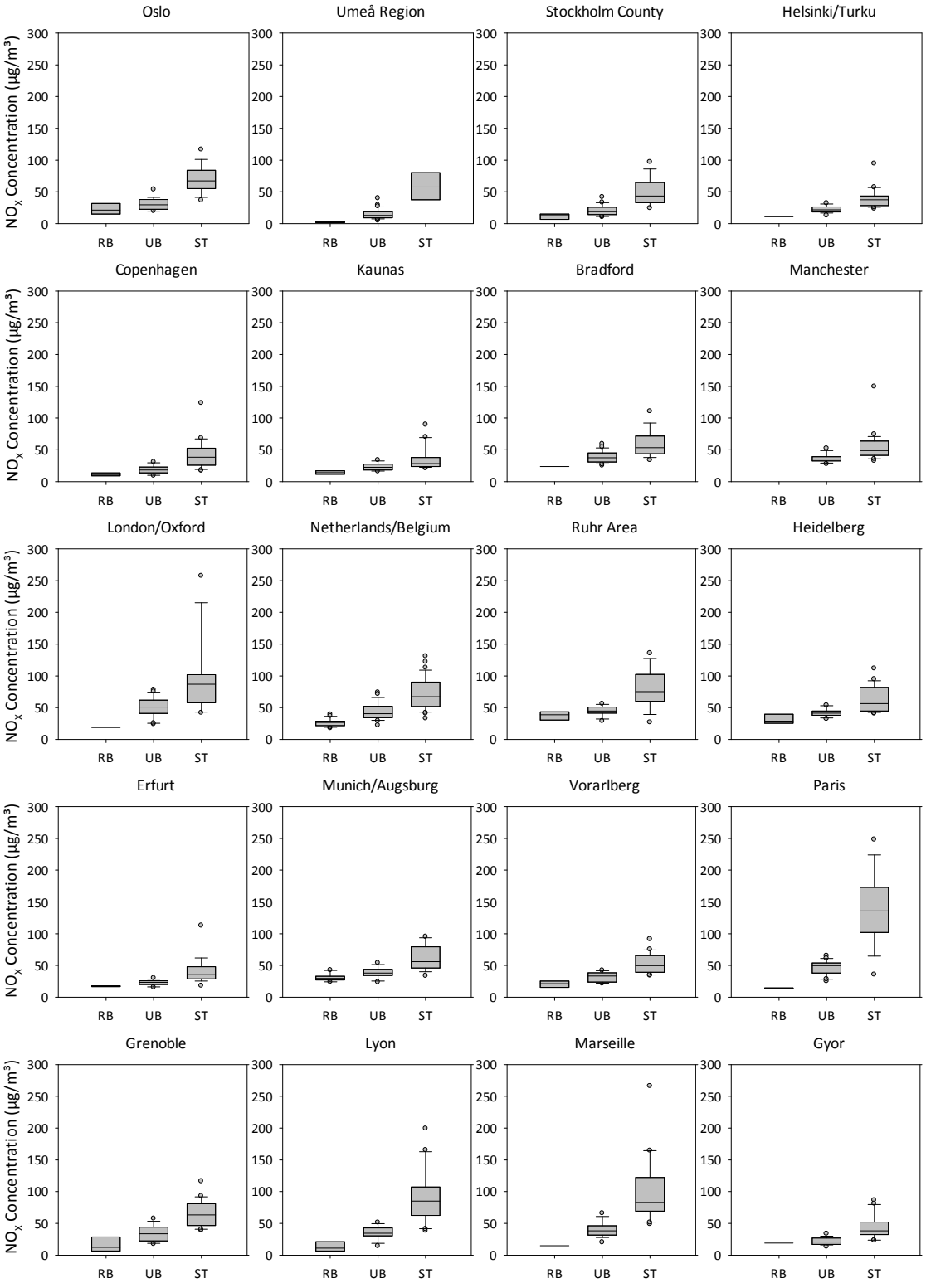


Figure 3: Annual averages of NO_2 concentrations by site type, for each study area (for measurement period please refer to Tabele 1). Median, 25^{th} and 75^{th} percentiles are shown in the box, whiskers indicate 10^{th} and 90^{th} percentiles and individual outliers are shown as points. RB = Regional Background, UB = Urban Background and ST = Street locations.


Oslo
Umeå Region
Stockholm County
Helsinki/Turku
Copenhagen
Kaunas
Bradford
Manchester
London/Oxford
Netherlands/Belgium
Ruhr Area
Heidelberg
Erfurt
Munich/Augsburg
Vorarlberg
Paris
Grenoble
Lyon
Marseille
Gyor
NOx Concentration (µg/m³)
0
50
100
150
200
250
300
RB
UB
ST

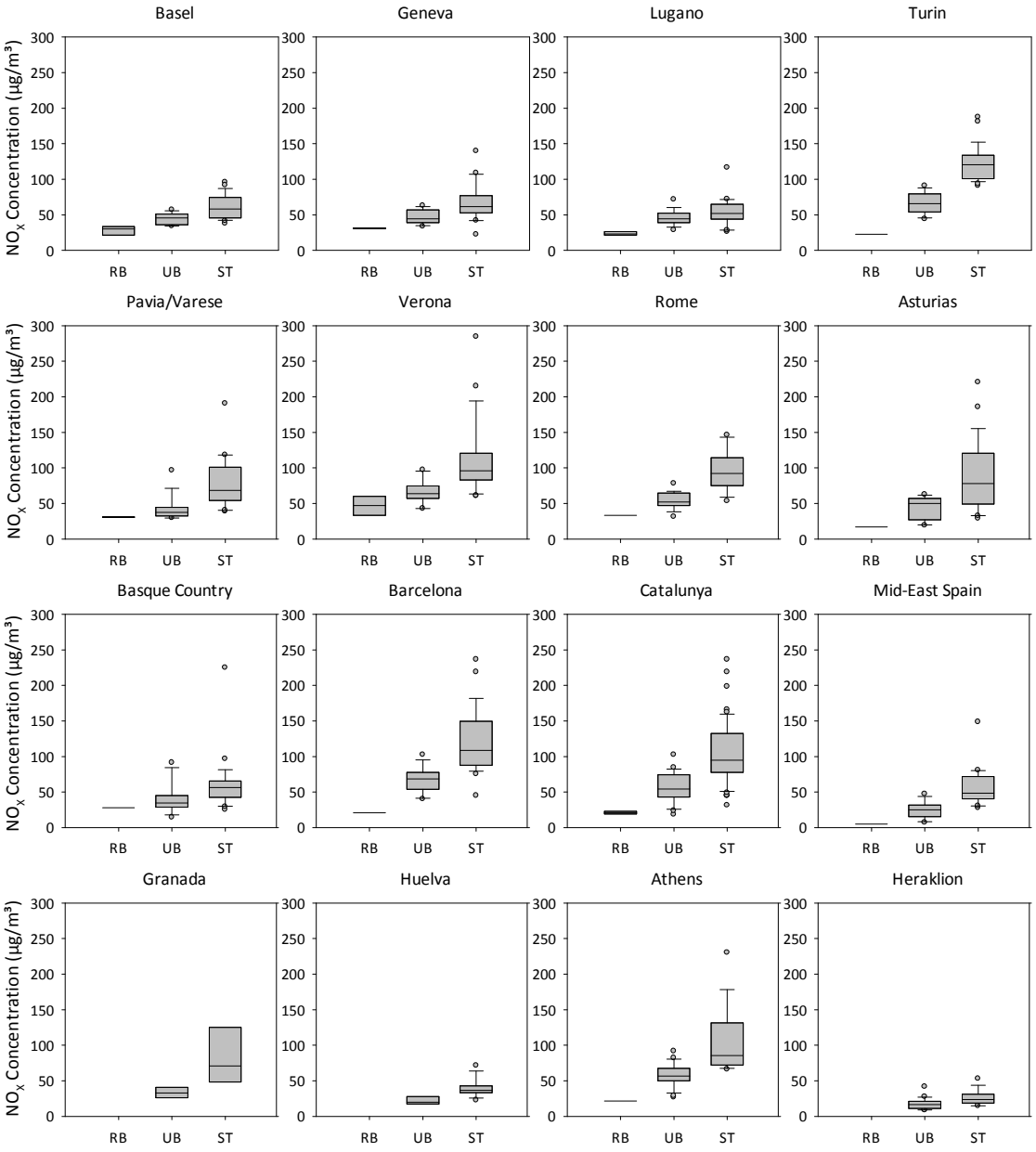


Figure 4: Annual averages of NO_x concentrations by site type, for each study area (for measurement period please refer to Tabele 1). Median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. RB = Regional Background, UB = Urban Background and ST = Street locations.

Table 1: Descriptive characteristics of all ESCAPE study areas

Country	Study area	Study area description; major cities	PM + NO_X?	Year	Measurement period	# of sites	Distribution over site types: [2] RB / UB / ST
Norway	Oslo	Oslo city	PM+ NO_x	1	05-02-2009 – 29-01-2010	40	4 / 18 / 18
Sweden	Umeå region	Vasterbotten county; Umeå, Skelleftea and Lycksele	NO_x	1	01-12-2008 - 11-07-2009	42	4 / 32 / 6
	Stockholm county	Stockholm county; Stockholm	PM+ NO_x	1	03-12-2008 – 01-12-2009	40	5 / 20 / 15
Finland	Helsinki/Turku	Two areas: Helsinki/Vantaa and Turku/Loimaa	PM+ NO_x	2	27-01-2010 – 26-01-2011	40	2 / 18 / 20
Denmark	Copenhagen	Copenhagen city and Hillerød	PM+ NO_x	2	19-11-2009 – 17-11-2010	41	6 / 13 / 22
Lithuania	Kaunas	Kaunas city	PM+ NO_x	2	20-01-2010 – 19-01-2011	40	5 / 13 / 22
United Kingdom	Bradford	Metropolitan borough of Bradford	NO_x	1	01-06-2009 – 15-12-2009	41	2 / 24 / 15
	Manchester	Greater Manchester urban area	PM+ NO_x	1	27-01-2009 – 20-01-2010	39	0 / 15 / 24
	London/Oxford	Thames valley; London, Oxford and smaller towns	PM+ NO_x	2	26-01-2010 – 18-01-2011	40	1 / 23 / 16
Netherlands/Belgium	Netherlands/Belgium	Entire country: Amsterdam, Rotterdam, Antwerp	PM+ NO_x	1	17-02-2009 – 19-02-2010	80	20 / 24 / 36
Germany	Ruhr area	Dortmund, Duisburg, Essen and smaller towns	PM+ NO_x	1	15-10-2008 – 12-10-2009	40	8 / 14 / 18
	Heidelberg	Heidelberg city and smaller surrounding towns	NO_x	1	07-04-2009 - 11-11-2009	43	3 / 16 / 24
	Erfurt	Erfurt city	NO_x	1	11-08-2009 - 16-12-2009	40	3 / 18 / 19
	Munich/Augsburg	Munich, Augsburg and smaller surrounding towns	PM+ NO_x	1	27-10-2008 – 05-11-2009	40	10 / 12 / 18
Austria	Vorarlberg	Cities and areas along the main valley of Vorarlberg	PM+ NO_x	2	03-03-2010 – 16-02-2011	40	5 / 11 / 24
France	Paris	Paris city and suburban areas	PM+ NO_x	2	04-01-2010 – 04-01-2011	40	4 / 20 / 16
	Grenoble	Grenoble city and suburban areas	NO_x	2	20-01-2010 - 07-07-2010	40	3 / 17 / 20
	Lyon	Lyon city and suburban areas	NO_x	2	20-01-2010 - 07-07-2010	40	3 / 17 / 20
	Marseille	Marseille city	NO_x	2	14-01-2010 - 24-06-2010	39	2 / 17 / 20
Hungary	Gyor	Gyor city and neighbouring villages	PM+ NO_x	2	22-02-2010 – 24-02-2011	40	1 / 19 / 20
Switzerland	Basel	Basel city and some surrounding smaller towns	NO_x	1	20-11-2008 - 10-06-2009	40	3 / 13 / 24
	Geneva	Geneva city and some surrounding smaller towns	NO_x	1	07-01-2009 - 03-07-2009	41	3 / 13 / 25
	Lugano	Lugano city and its neighboring communities	PM+ NO_x	1	02-03-2009 – 10-03-2010	42	4 / 16 / 22
Italy	Turin	Turin city and five smaller municipalities	PM+ NO_x	2	01-02-2010 – 25-01-2011	40	1 / 13 / 26
	Pavia/Varese	Cities of Pavia, Varese and surrounding areas	NO_x	2	08-02-2010 - 14-06-2010	40	3 / 14 / 23
	Verona	City of Verona and surrounding areas	NO_x	2	20-01-2010 - 22-06-2010	40	3 / 14 / 23
	Rome	Rome city	PM+ NO_x	2	27-01-2010 – 26-01-2011	40	2 / 19 / 19
Spain	Asturias	North part of Asturias and Oviedo region: Oviedo and Avilés	NO_x	1	17-02-2009 - 22-06-2009	40	2 / 13 / 25
	Basque country	Galdakao, Gipuzkoa and San Sebastián areas; many small towns	NO_x	1	03-02-2009 - 15-07-2009	39	2 / 12 / 25
	Barcelona	Barcelona city	PM+ NO_x	1	14-01-2009 – 14-01-2010	40	1 / 13 / 26
	Catalunya	Three areas around Barcelona, Girona, Sabadell	PM+ NO_x	1	14-01-2009 – 14-01-2010	80	5 / 23 / 52
	Mid East Spain	Valencia region and Albacete city	NO_x	1	17-02-2009 - 23-07-2009	38	2 / 13 / 23
	Granada	Granada city and smaller towns around Granada and Loja	NO_x	1	17-03-2009 - 15-09-2009	14	0 / 7 / 7
	Huelva	Hulvea city	NO_x	1	17-03-2009 - 15-09-2009	24	0 / 8 / 16
Greece	Athens [1]	Greater Athens area, 16 municipalities; Athens	PM+ NO_x	2	21-04-2010 – 27-04-2011	40	1 / 22 / 17
	Heraklion [1]	Heraklion prefecture; Heraklion	PM+ NO_x	1	18-02-2009 – 16-02-2010	40	0 / 21 / 19

[1] PM+NOx areas: dates refer to the period when the reference site was operated. NOx only areas: dates refer to the start of first and end of third measurement period.

[2] RB = regional background / UB = urban background / ST = street site

Table 2: Detection limits and precision for NO_2 and NO_x measurements

	Field blanks							Field duplicates		
	Number field blanks	Average field blank (µg/m³)		Detection limit (µg/m³)		N samples below the detection limit (total number of valid samples)		Number duplicates	CV (%)	
Study area	N	NO_2	NOx	NO_2	NOx	NO_2	NOx	N	NO_2	NOx
Oslo	18	0.0	0.4	2.5	2.8	1 (123)	0 (123)	28	23.1	11.5
Umeå Region	6	0.1	0.7	0.6	1.2	0 (124)	7 (124)	6	4.5	4.8
Stockholm County	22	0.1	0.5	1.1	2.4	2 (143)	0 (143)	22	9.5	7.5
Helsinki/Turku	23	0.1	0.2	0.6	1.7	0 (154)	0 (154)	24	4.9	6.2**
Copenhagen	20	0.3	-0.2	1.2	3.1	0 (143)	0 (143)	20	5.6	7.6
Kaunas*	24	0.1	0.8	0.6*	5.7*	0 (146)	3 (146)	24	8.5	10.4**
Bradford	5	0.5	0.9	1.1	2.4	0 (112)	0 (112)	6	12.0	1.9
Manchester	12	0.5	1.5	1.1	5.3	1 (116)	0 (116)	12	4.9	4.4
London/Oxford	23	0.3	1.3	1.3	5.5	0 (131)	0 (131)	23	5.4	5.7
Netherlands / Belgium	20	0.2	0.5	1.3	3.6	0 (263)	0 (263)	20	7.1	4.7
Ruhr Area	15	0.2	0.4	0.8	2.5	0 (120)	0 (120)	23	4.5	4.5
Heidelberg	8	0.3	1.1	0.7	2.3	0 (127)	0 (127)	10	9.7	3.2
Erfurt	12	0.2	0.0	0.8	2.6	0 (118)	0 (118)	12	8.8	3.8
Munich	15	0.3	0.9	1.0*	4.6*	0 (142)	0 (142)	16	3.0	2.9
Vorarlberg	25	0.6	2.4	1.2	6.7	1 (144)	1 (144)	25	9.3	6.6
Paris	22	0.3	2.1	1.2	6.6	0 (141)	0 (141)	22	8.9	7.1
Grenoble	12	0.1	0.5	0.8	3.1	0 (120)	0 (120)	12	19.5	7.5
Lyon	12	0.2	0.9	0.5	3.6	0 (117)	0 (117)	12	7.9	6.3
Marseille	12	0.1	0.1	0.6	1.6	0 (114)	0 (114)	17	4.5	4.3
Gyor	25	0.3	0.6	1.3	2.5	0 (145)	0 (145)	24	10.7	13.2
Basel	8	0.4	0.8	0.7	5.0	0 (120)	0 (120)	8	4.9	5.6
Geneva	10	0.1	1.1	1.0	8.3	0 (121)	0 (121)	8	5.6	4.1
Lugano	16	0.5	1.5	1.2*	9.5*	0 (137)	4 (137)	23	7.7	5.1
Turin	24	0.3	1.7	1.4*	10.0*	0 (144)	36 (144)	24	9.6	10.0**
Pavia/Varese	12	0.8	1.2	2.2	3.2	0 (120)	0 (120)	12	9.3	5.2
Verona	11	0.3	0.3	0.5	2.2	0 (120)	0 (120)	13	7.2	2.5
Rome	24	0.2	0.4	0.5	2.0	0 (144)	0 (144)	25	9.0	4.3
Asturias:***										
Asturias region	12	0.0	0.0	0.9	1.9	0 (45)	0 (45)	12	5.1	4.5
Oviedo	12	0.4	3.8	1.6	17.2	0 (68)	8 (68)	12	5.6	2.3
Basque Country:***										
Bilbao	12	0.2	0.2	1.0	3.0	0 (40)	0 (40)	12	6.4	3.3
San Sebastian	11	0.0	0.4	0.8	1.3	0 (71)	0 (71)	9	4.6	2.9
Barcelona	23	0.5	3.7	3.3*	28.1*	1 (142)	6 (142)	27	7.7	7.8
Catalunya	23	0.5	3.7	3.3*	28.1*	1 (309)	39 (309)	27	7.7	7.8
Mid-East Spain:***										
Albacete	12	0.4	3.1	1.9	9.4	1 (51)	12 (51)	10	9.9	14.2
Valencia	10	0.1	0.1	0.4	1.0	1 (51)	1 (51)	11	11.9**	3.9
Granada	3	0.5	0.7	1.1	1.5	0 (29)	1 (29)	3	35.6	5.5
Huelva	2	-0.2	0.0	0.3	2.7	0 (67)	0 (67)	3	19.5	13.4
Athens	23	0.1	0.3	0.8	1.3	0 (143)	1 (143)	23	10.9	5.6
Heraklion	21	0.5	0.7	2.7	3.5	2 (140)	1 (140)	13	7.8	5.6

Detection limit calculated as three times the standard deviation of field blanks. CV= coefficient of variation calculated from field duplicates.

* Detection limit (DL) affected by one blank for which no explanation was found. Without this one blank, NO_X DL and number of samples below DL become 2.0 µg/m³ and 0 (Munich): 6.7 µg/m³ and 27 (Turin); 2.7 µg/m³ and 0 (Kaunas); 7.2 µg/m³ and 0 (Barcelona); 7.2 µg/m³ and 0 (Catalunya); 2.8 µg/m³ and 0 (Lugano);

** CV affected by one poor duplicate, for which no explanation was found. Without this duplicate CV becomes 3.8% for NO_X (Helsinki); 5.4% for NO_X (Kaunas); 3.1% for NO_X (Turin); 4.7% for NO_2 (Valencia);

*** in three subareas different teams performed fieldwork, field duplicates / blanks treated analysed separately

Table 3: Ratios between regional background and urban background concentrations, and between street and urban background concentrations, for all study areas.

	NO_2		NO_x	
StudyArea	Ratio Regional / Urban background	Ratio Street / Urban background	Ratio Regional / Urban background	Ratio Street / Urban background
Oslo	0.71*	2.09**	0.73*	2.28**
Umeå Region	0.26**	3.16**	0.18**	4.24**
Stockholm County	0.49**	1.86**	0.50**	2.35**
Helsinki/Turku	0.59**	1.55**	0.48**	1.71**
Copenhagen	0.69**	1.72**	0.64**	2.14**
Kaunas	0.66**	1.33**	0.62**	1.43**
Bradford	0.71**	1.22**	0.63**	1.52**
Manchester	--	1.32**	--	1.42**
London/Oxford	0.24**	1.50**	0.39**	1.81**
Netherlands/Belgium	0.64**	1.36**	0.62**	1.63**
Ruhr Area	0.90	1.41**	0.88	1.70**
Heidelberg	0.75*	1.34**	0.74*	1.44**
Erfurt	0.73*	1.48**	0.75	1.65**
Munich/Augsburg	0.79**	1.38**	0.81*	1.56**
Vorarlberg	0.68**	1.38**	0.65**	1.63**
Paris	0.30**	1.98**	0.30**	2.77**
Grenoble	0.44**	1.66**	0.41**	1.94**
Lyon	0.47**	1.93**	0.34**	2.51**
Marseille	0.46**	1.78**	0.38**	2.39**
Gyor	0.91	1.46**	0.89	1.87**
Basel	0.72**	1.24**	0.64**	1.35**
Geneva	0.71**	1.35**	0.67*	1.36**
Lugano	0.56**	1.09	0.53**	1.14
Turin	0.43**	1.71**	0.35**	1.85**
Pavia/Varese	0.75	1.59**	0.78	1.79**
Verona	0.65**	1.44**	0.70	1.63**
Rome	0.55**	1.48**	0.60**	1.71**
Asturias	0.42**	1.58**	0.41**	1.84**
Basque Country	0.72	1.38**	0.75	1.49**
Barcelona	0.33**	1.52**	0.32**	1.76**
Catalunya	0.42**	1.62**	0.41**	1.89**
Mid-East Spain	0.24**	2.12**	0.11**	2.43**
Granada	--	2.30**	--	2.34**
Huelva	--	1.75**	--	1.81**
Athens	0.44**	1.45**	0.38**	1.80**
Heraklion	--	1.60**	--	1.56**

*=significant difference between the site types on $p<0.10$ level

**=significant on $p<0.05$ level

Table 4: Correlation and ratio between NO_2 and NO_x (R-squared).

	Study area	Correlation between NO_2 and NO_x (R^2)*	Ratio NO_2/NO_x			
			All sites	Regional background	Urban background	Street
1	Oslo	0.96	0.51	0.53	0.54	0.49
2	Umeå Region	0.97	0.57	0.81	0.56	0.42
3	Stockholm County	0.93	0.58	0.62	0.63	0.50
4	Helsinki/Turku	0.91	0.64	0.82	0.66	0.61
5	Copenhagen	0.94	0.65	0.78	0.72	0.58
6	Kaunas	0.91	0.62	0.68	0.64	0.60
7	Bradford	0.88	0.60	0.72	0.64	0.52
8	Manchester	0.91	0.58	--	0.60	0.56
9	London/Oxford	0.93	0.58	0.39	0.63	0.53
10	Netherlands/Belgium	0.94	0.63	0.70	0.68	0.56
11	Ruhr Area	0.97	0.59	0.66	0.64	0.53
12	Heidelberg	0.97	0.62	0.66	0.65	0.60
13	Erfurt	0.92	0.65	0.66	0.68	0.62
14	Munich/Augsburg	0.95	0.59	0.60	0.62	0.55
15	Vorarlberg	0.83	0.55	0.64	0.60	0.51
16	Paris	0.93	0.57	0.63	0.64	0.46
17	Grenoble	0.93	0.60	0.70	0.64	0.55
18	Lyon	0.96	0.64	0.95	0.71	0.54
19	Marseille	0.94	0.58	0.80	0.66	0.49
20	Gyor	0.90	0.55	0.63	0.62	0.48
21	Basel	0.94	0.60	0.70	0.62	0.57
22	Geneva	0.88	0.53	0.56	0.53	0.53
23	Lugano	0.83	0.62	0.66	0.62	0.61
24	Turin	0.94	0.54	0.69	0.56	0.52
25	Pavia/Varese	0.96	0.53	0.55	0.57	0.51
26	Verona	0.95	0.47	0.47	0.51	0.45
27	Rome	0.94	0.60	0.59	0.65	0.56
28	Asturias	0.95	0.49	0.55	0.54	0.46
29	Basque Country	0.92	0.50	0.51	0.53	0.49
30	Barcelona	0.93	0.60	0.66	0.65	0.57
31	Catalunya	0.92	0.59	0.66	0.64	0.56
32	Mid-East Spain	0.98	0.71	1.93**	0.70	0.61
33	Granada	0.95	0.61	--	0.61	0.61
34	Huelva	0.97	0.65	--	0.67	0.64
35	Athens	0.81	0.51	0.62	0.55	0.44
36	Heraklion	0.92	0.72	--	0.71	0.73

Ratios between NO_2 and NO_x for all sites, regional background sites and street sites.
*All p-values were below <0.0001.
**=This value is different because in this particular case the ratio was only based on 2 regional background sites of which one had a negative NO concentration and hence a NO_x concentration which was lower than NO_2.